The Chemistry on Diterpenoids in 1980 (Commemoration Issue Dedicated to Professor Yuzo Inouye on the Occasion of his Retirement)

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REVIEW

The Chemistry on Diterpenoids in 1980

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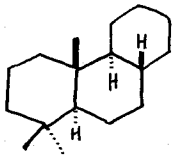
I. INTRODUCTION

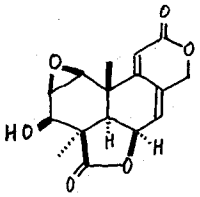
This is one of a series of our annual reviews on diterpenoid chemistry. The style is changed from that in the previous reviews. The following abbreviations are used. [CN]: common name; [NS]: natural source; [REF]: reference number; [NC]: notes and comments.

II. PODOCARPANE DERIVATIVES



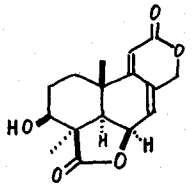
Podocarpane

1) Isolation and Structure Determination



1

[CN] wentilactone A
[NS] *Aspergills wentii*
[REF] 1
[NC] X-ray analysis



2

[CN] wentilactone B
[NS] *Aspergills wentii*
[REF] 1

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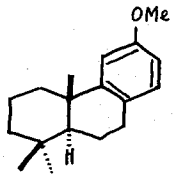
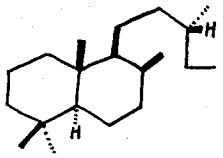
2) Synthesis and Reaction



3

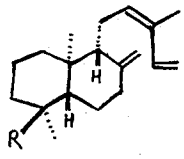
[REF] 2
[NC] biogenetic-type cyclization

III. LABDANE DERIVATIVES

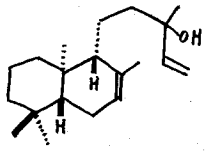


Labdane

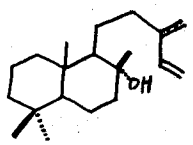
1) Isolation and Structure Determination



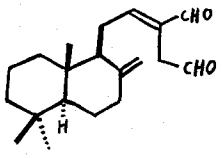
4 R=Me
5 R=CHO
6 R=CO_2H
[NS] *Helianthus* species
[REF] 3



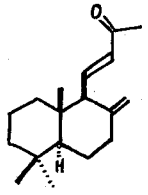
7
[NS] *Helichrysum* species
[REF] 4



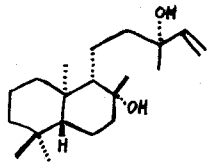
8
[NS] *Smallanthus fruticosus*
[REF] 5



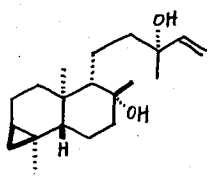
9
[NS] *Alpinia speciosa*
[REF] 6



10
[NS] *Alpinia speciosa*
[REF] 6

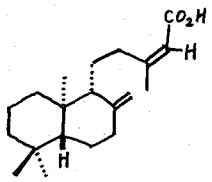


11
[NS] *Gnaphalium undulatum*
[REF] 7



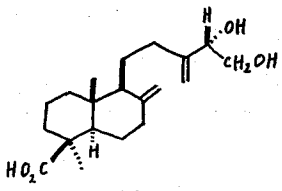
12

[NS] *Gnaphalium undulatum*
[REF] 7



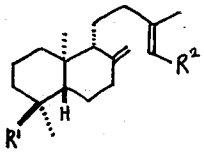
13

[NS] *Morithamnus crassus*
[REF] 8

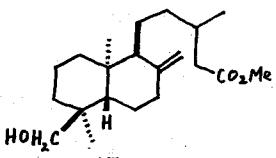


14

[NS] *Juniperus communis*
[REF] 9

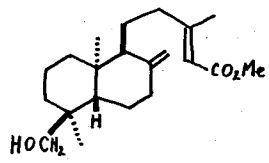


15 $R^1=CO_2H$, $R^2=CH_2OH$
[CN] visidic acid A
16 $R^1=CO_2H$, $R^2=CH_2OAc$
[CN] visidic acid B
[NS] *Chrysothammus viscidiflorus*
[REF] 10



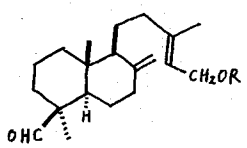
17

[NS] rosin of "Brazil capal"
[REF] 11

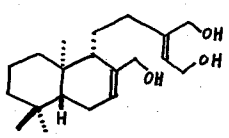


18

[NS] rosin of "Brazil capal"
[REF] 11

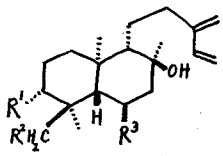


19 R=H
20 R=D-xylose
[NS] *Thujopsis dolabrata*
[REF] 12

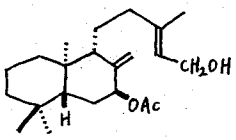


21

[NS] *Achyrocline alata*
[REF] 13

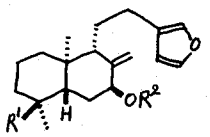


22 $R^1=H$, $R^2=OH$, $R^3=OAc$
23 $R^1=H$, $R^2=OAc$, $R^3=OH$
24 $R^1=OH$, $R^2=H$, $R^3=OAc$
[NS] *Sideritis foetens*
[REF] 14

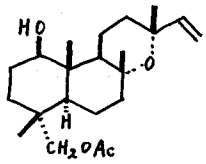


25

[NS] *Austroeupatorium chaparense*
[REF] 15



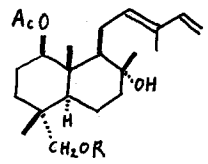
26 $R^1=Me$, $R^2=Ac$
27 $R^1=CHO$, $R^2=Ac$
28 $R^1=CH_2OAc$, $R^2=Ac$
29 $R^1=CH_2OH$, $R^2=Ac$
[NS] *Austroeupatorium chaparense*
[REF] 15



30

[NS] *Aristequietia buddleaefolia*

[REF] 16



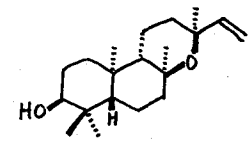
31 R=$CO(CH_2)_{18}Me$

32 R=$COCH{=}CHC_6H_4OH(p)$

33 R=$COCH{=}CHC_6H_4OAc(p)$

[NS] *Aristequietia buddleaefolia*

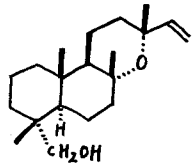
[REF] 16



34

[NS] *Subtribus espeletinae*

[REF] 17

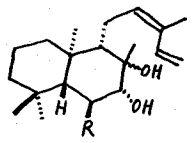


35

[CN] jhanol

[NS] *Stevia rebaudiana*

[REF] 18



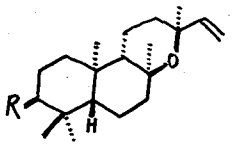
36 R=OH

[CN] austroinulin

37 R=OAc

[NS] *Stevia rebaudiana*

[REF] 18

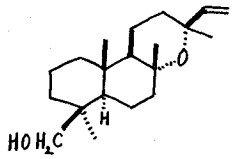


34 R=OH

38 R=H

[NS] *Libanothamnus spectabilis*

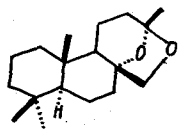
[REF] 19



39

[NS] *Baccharis tola*

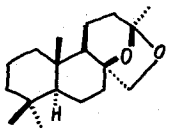
[REF] 20



40

[NS] *Pinus monticola*

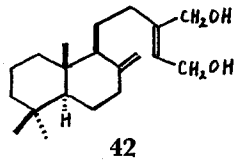
[REF] 21



41

[NS] *Pinus monticola*

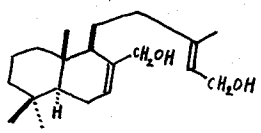
[REF] 21



42

[NS] *Ceroplastes ceriferus*

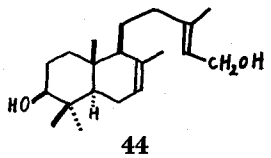
[REF] 22



43

[NS] *Ceroplastes ceriferus*

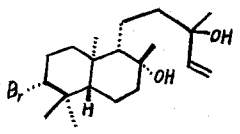
[REF] 22



44

[NS] *Ceroplastes ceriferus*

[REF] 22

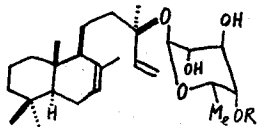


45

[CN] isoconcinndiol

[NS] *Laurencia snyderae var. guadalupensis*

[REF] 23

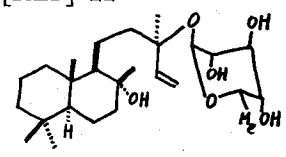


46 R=H

47 R=Ac

[NS] *Aster spathulifolius*

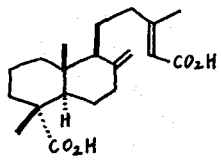
[REF] 24



48

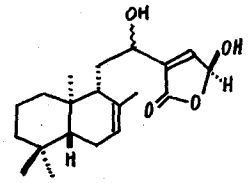
[NS] *Aster spathulifolius*

[REF] 24



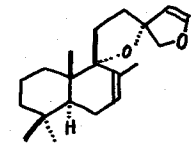
49

[CN] dehydropinifolic acid
[NS] *Pinus silvestris*
[REF] 25



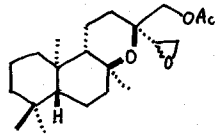
50

[NS] *Acritopappus* species
[REF] 26



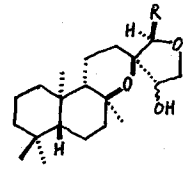
51

[NS] *Solidago* species
[REF] 27

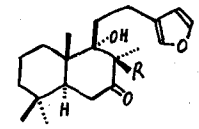


52

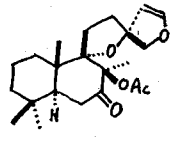
[NS] *Schkuhria* species
[REF] 28



53 R=OH
54 R=OAc
[NS] *Schkuhria* species
[REF] 28

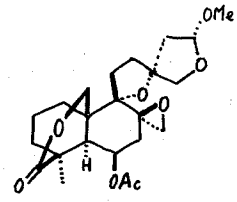


55 R=H
[CN] hispanolone
56 R=OAc
[CN] galeopsin
[NS] *Galeopsis angustifolia*
[REF] 29



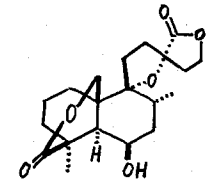
57

[CN] pregaleopsin
[NS] *Galeopsis angustifolia*
[REF] 29



58

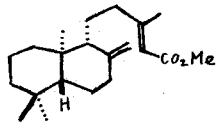
[CN] methoxynepetaefolin
[NS] *Leonotis nepetaefolia*
[REF] 30
[NC] X-ray analysis



59

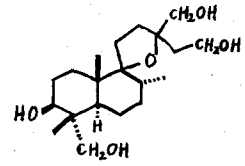
[CN] nepetaefolinol
[NS] *Leonotis nepetaefolia*
[REF] 30

2) Synthesis and Reaction



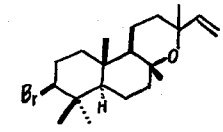
60

[CN] methyl copalate
[REF] 31
[NC] synthesis of debromoisoaplysin-20 and its C–13 epimer from **60**



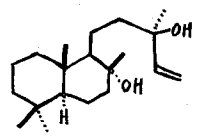
61

[CN] lagochilin
[REF] 32
[NC] ·acetylation of **61**
·NMR studies



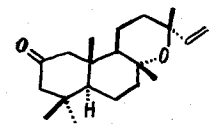
62

[REF] 33
[NC] brominative cyclization of geranyl-linalool



63

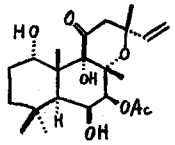
[CN] sclareol
[REF] 34~37
[NC] oxidation of **63** by a chromium mixture



64

[CN] 2-oxomanoyl oxide
[REF] 38
[NC] chemical conversion

3) Miscellaneous Section

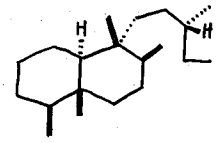


65

[CN] forskolin
[NS] *Coleus* and *Plectranthus* species
[REF] 39
[NC] TLC and GLC assay methods for the presence of **65**

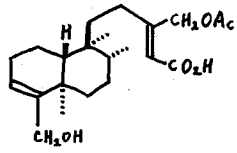
Additional references
[REF] 40
[NC] Inheritance of labdanoid producing ability in *Nicotiana Tabacum*.
[REF] 41
[NC] A review article on the chemistry of the *Compositae* diterpenoids.

IV. CLERODANE DERIVATIVES



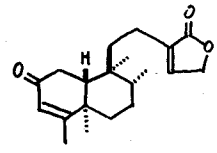
Clerodane

1) Isolation and Structure Determination



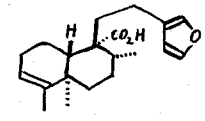
66

[NS] *Acritopappus* species
[REF] 26



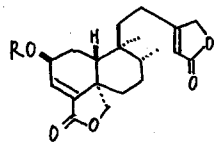
67

[NS] *Acritopappus* species
[REF] 26

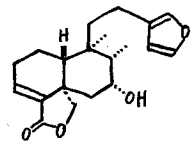


68

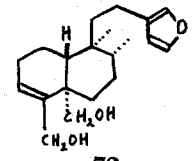
[NS] *Solidago* species
[REF] 27



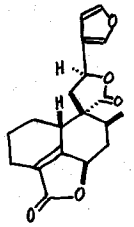
69 R=H
[CN] articulin
70 R=AC
[CN] articulinacetate
[NS] *Baccharis articulata*
[REF] 42



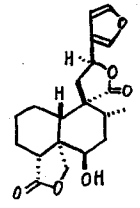
71
[CN] 1-deoxybacrispin
[NS] *Baccharis crispa*
[REF] 43



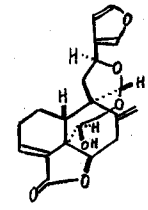
72
[CN] hautriwaic acid
[NS] *Baccharis crispa*
[REF] 43



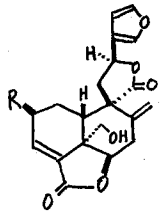
73
[CN] teuchamaedryn A
[NS] *Teucrium chamaedrys*
[REF] 44



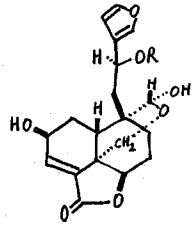
74
[CN] teuchamaedryn B
[NS] *Teucrium chamaedrys*
[REF] 44



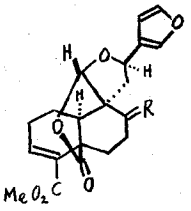
75
[CN] Plaunol A
[NS] *Croton sublyratus*
[REF] 45
[NC] X-ray analysis



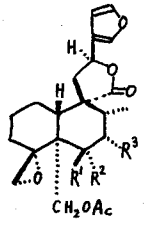
76 R=H
77 R=OH
[CN] plaunol C
[NS] *Croton sublyratus*
[REF] 45



78 R=H
[CN] plaunol D
79 R=Ac
[CN] plaunol E
[NS] *Croton sublyratus*
[REF] 45

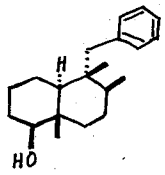


80 R=β-Me, α-H
[CN] dihydrocroverin
[NC] anti-peptic ulcer activity
81 R=CH_2
[CN] croverin
[NS] *Croton verreauxii*
[REF] 46



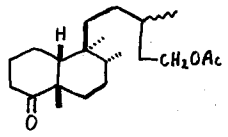
82 R^1, R^2=O, R^3=OAc
[CN] capitatin
83 R^1=OAc, R^2=H, R^3=OH
[CN] teucapitatin
[NS] *Teucrium capitatum*
[REF] 47

2) Synthesis and Reaction



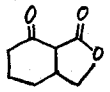
84

[REF] 48
[NC] synthesis



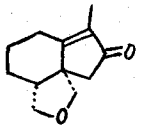
85

[REF] 49
[NC] photoisomerization



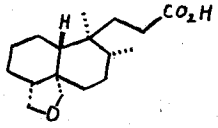
86

[REF] 50
[NC] synthesis



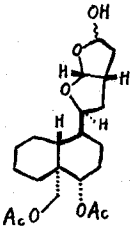
87

[REF] 51
[NC] synthesis



88

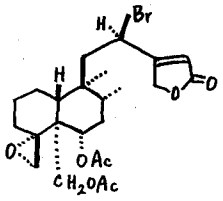
[REF] 52
[NC] synthesis



89

[REF] 53
[NC] synthesis

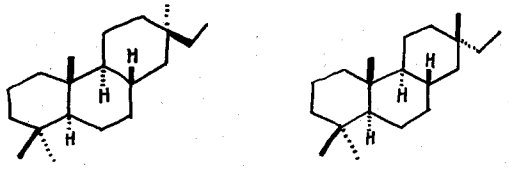
3) Miscellaneous Section



90

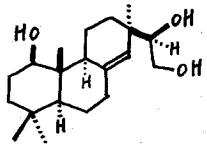
[CN] 12(R)-bromoajugarin-I
[REF] 54
[NC] ·bromination ·X-ray analysis

V. PIMARANE AND ISOPIMARANE DERIVATIVES



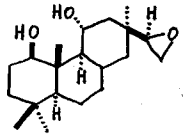
Pimarane and Isopimarane

1) Isolation and Structure Determination



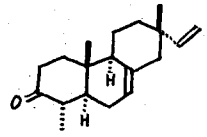
91

[CN] leucophleol
[NS] *Acacia leucophloea*
[REF] 55



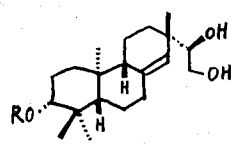
92

[CN] leucophleoxol
[NS] *Acacia leucophloea*
[REF] 55

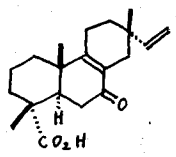


93

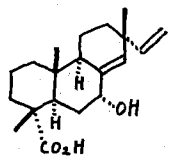
[NS] *Acremonium luzulae*
[REF] 56



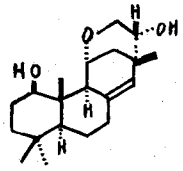
94 R=H
[CN] darutigenol
95 R=β-D-glucose
[CN] darutoside
[NS] *Sigesbeckia orientalis*
[REF] 57



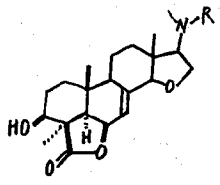
96
[NS] *Juniperus communis*
[REF] 9



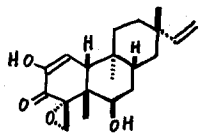
97
[NS] *Juniperus communis*
[REF] 9



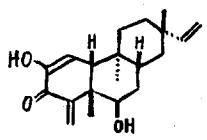
98
[NS] *Acacia leucophloea*
[REF] 58



99 R=Me
[CN] icaceine
100 R=H
[CN] de-N-methylicaceine
[NS] *Icacina guesfeldtii*
[REF] 59

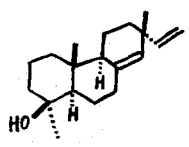


101
[CN] oxidopanamensin
[NS] *Rondeletia panamensis*
[REF] 60

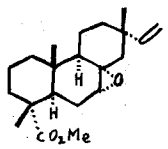


102
[CN] panamensin
[NS] *Rondeletia panamensis*
[REF] 60

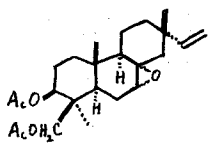
2) Synthesis and Reaction



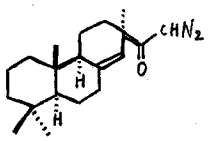
103
[REF] 61
[NC] synthesis of **103** and its C–4 epimer



104
[REF] 62
[NC] rearrangement of **104** with BF_3–Et_2O



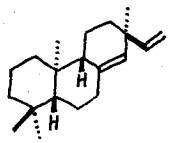
105
[REF] 63
[NC] acid catalyzed ring cleavage of **105**



106

[REF] 64
[NC] acid catalyzed clyclization of **106**

3) Miscellaneous Section

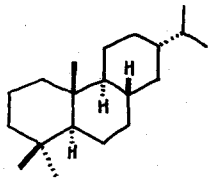


107

[CN] *ent*-sandaracopimaradiene
[REF] 65
[NC] biosynthesis

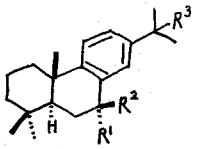
Additional references
[REF] 66–68
[NC] ^{13}C NMR studies

VI. ABIETANE DERIVATIVES

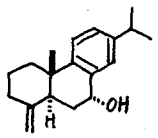


Abietane

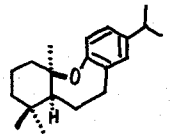
1) Isolation and Structure Determination



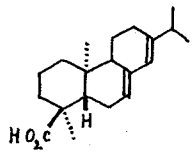
108 $R^1=R^2=H$, $R^3=OH$
109 $R^1=OH$, $R^2=R^3=H$
110 R^1, $R^2=O$, $R^3=H$
[NS] *Pinus monticola*
[REF] 21



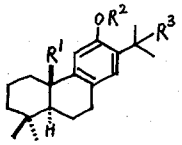
111
[NS] *Pinus monticola*
[REF] 21



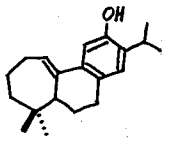
112
[NS] *Pinus monticola*
[REF] 21



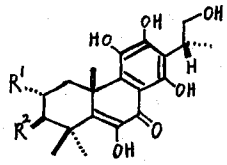
113
[NS] *solidago* species
[REF] 27



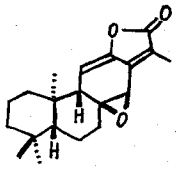
114 R^1=Me, R^2=H, R^3=OH
115 R^1=CO_2H, R^2=Me, R^3=H
116 R^1=CHO, R^2=R^3=H
[NS] *Chamaecyparis pisifera*
[REF] 69



117
[CN] pisiferin
[NS] *Chamaecyparis pisifera*
[REF] 69

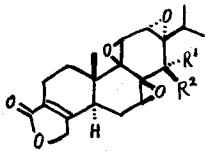


118 R^1=R^2=H
[CN] coleon C
119 R^1=H, R^2=OAc
[CN] coleon H
120 R^1=OCHO, R^2=H
[NS] *Solenostemon monostachys*
[REF] 70

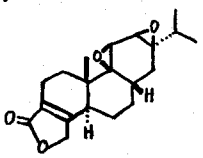


121
[CN] jolkinolide A
[NS] *Euphorbia huachangana*
[REF] 71

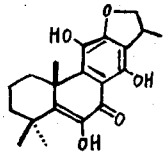
2) Synthesis and Reaction



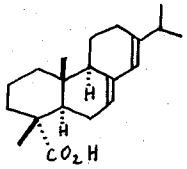
122 R^1=H, R^2=OH
[CN] triptolide
123 R^1, R^2=O
[CN] triptonide
[REF] 72, 73
[NS] synthesis



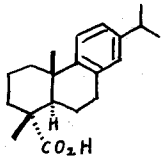
124
[CN] stemolide
[REF] 74
[NC] synthesis



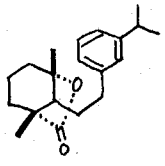
125
[CN] lycoxanthol
[REF] 75
[NC] synthetic studies of **125**



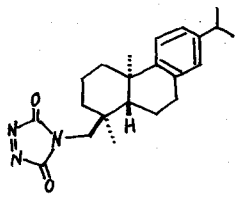
126
[CN] abietic acid
[REF] 76
[NC] Formation of coeon A skeleton from **126**



127
[REF] 77
[NC] synthesis of carotene derivative from **127**

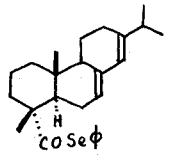


128
[REF] 78
[NC] synthesis and structure determination



129

[REF] 79
[NC] asymmetric Diels-Alder reaction of **129**



130

[REF] 80
[NC] reduction of **130** with Bu_3SnH

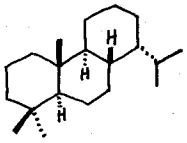
3) Miscellaneous Section

126

[REF] 81
[NC] allergic constituents of some plants

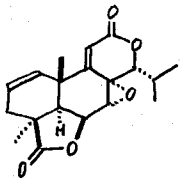
Additional reference
[REF] 82
[NC] studies of hypocholesterolemic activity of abietamide derivatives

VII. TOTARANE DERIVATIVES



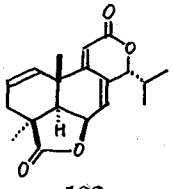
Totarane

1) Isolation and Structure Determination



131

[CN] milanjilactone A
[NS] *Podocarpus milanjianus*
[REF] 83



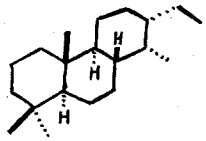
132

[CN] milanjilactone B
[NS] *podocarpus milanjianus*
[REF] 83

2) Miscellaneous Section

Additional reference
[REF] 84
[NC] ^{13}C NMR studies

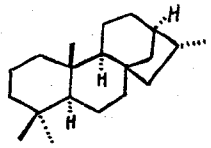
VIII. CASSANE DERIVATIVES



Cassane

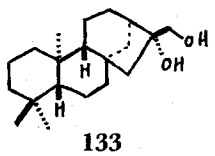
No report

IX. KAURANE DERIVATIVES



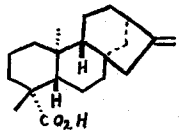
Kaurane

1) Isolation and Structure Determination



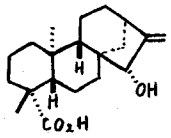
133

[NS] *Morithamnus crassus*
[REF] 8



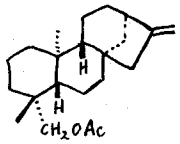
134

[NS] *Helianthus niveus* and others
[REF] 3, 4, 7, 19, 85, 86, 87



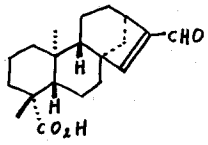
135

[NS] *Helianthus niveus* and others
[REF] 7, 85; *cf.* 86



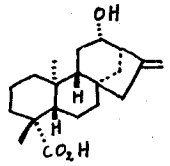
136

[NS] *Espeletiopsis* species
[REF] 88



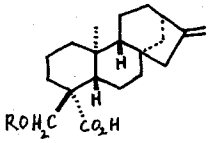
137

[NS] *Espeletiopsis* species
[REF] 88



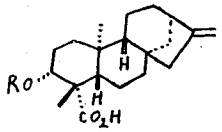
138

[NS] *Smallanthus uvedalis*
[REF] 87



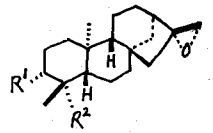
139 R=angeloyl
140 R=senecionly
141 R=isovaleryl

[NS] *Smallanthus uvedalis*
[REF] 87



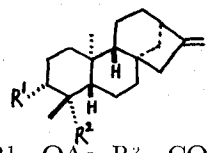
142 R=angeloyl
143 R=senecionyl
144 R=isovaleryl

[NS] *Smallanthus uvedalis*
[REF] 87

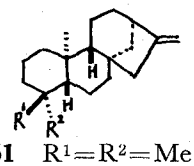


145 R^1=H, R^2=CH_2O-angeloyl
146 R^1=O-angeloyl, R^2=Me
147 R^1=O-angeloyl, R^2=CHO

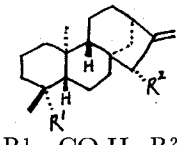
[NS] *Smallanthus uvedalis*
[REF] 87



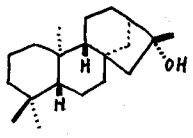
148 R^1=OAc, R^2=CO_2H
149 R^1=OH, R^2=CH_2OH
150 R^1=OH, R^2=CO_2H
[NS] *Stachys lanata*
[REF] 89



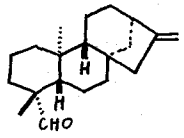
151 R^1=R^2=Me
152 R^1=Me, R^2=H
153 R^1=H, R^2=Me
[NS] *Libanothamnus granatesianus*
[REF] 19



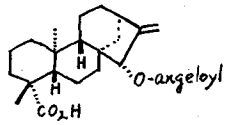
154 R^1=CO_2H, R^2=OAc
155 R^1=CO_2H, R^2=O isovaleryl
156 R^1=CH_2OH, R^2=H
[NS] *Libanothamnus granatesianus*
[REF] 19



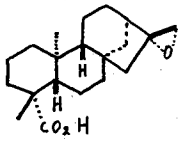
157
[NS] *Helichrysum* species
[REF] 4



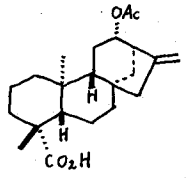
158
[NS] *Gnaphalium undulatum*
[REF] 7



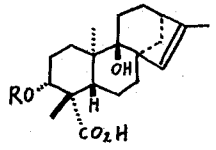
159
[NS] *Smallanthus fruticosus* and others
[REF] 3, 5, 27



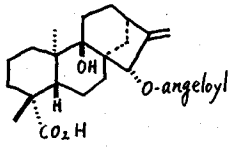
160
[NS] *Smallanthus fruticosus*
[REF] 5



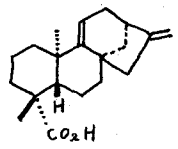
161
[NS] *Helianthus* species
[REF] 3



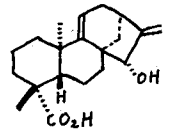
162 R=isovaleryl
163 R=isobutyl
[NS] *Polymnia canadensis*
[REF] 90



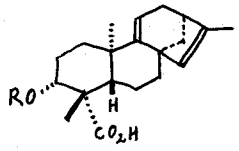
164
[NS] *Steiractinia mollis*
[REF] 91



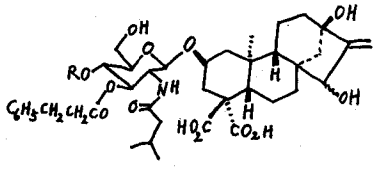
165
[NS] *Steiractinia mollis* and others
[REF] 3, 7, 19, 91, 92



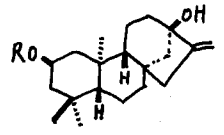
166
[NS] *Smallanthus fruticosus*
[REF] 5



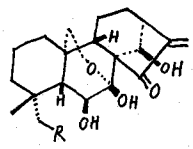
167 R=isovaleryl
168 R=isobutyl
169 R=tigl
[NS] *Polymnia canadensis*
[REF] 90



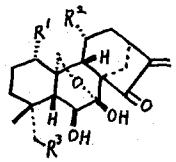
170 R=H
[CN] wedeloside
171 R=L-rhamnopyranosyl
[NS] *Wedelia asperrima*
[REF] 93



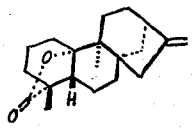
172 R=β-D-glucosyl
[NS] *Lindsaea chienii*
[REF] 94



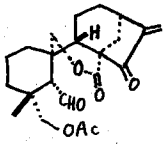
173 R=H
174 R=OAc
[CN] longikaurin A (173) and B (174)
[NS] *Rabdosia longituba*
[REF] 95



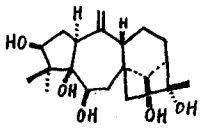
175 R^1=OH, R^2=R^3=H
176 R^1=OAc, R^2=R^3=H
177 R^1=OH, R^2=H, R^3=OAc
178 R^1=R^3=OAc, R^2=H
179 R^1=R^2=OH, R^3=H
[CN] effusanin A–E
[NS] *Rabdosia effusa*
[REF] 96



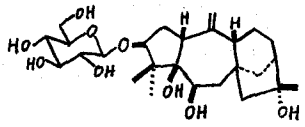
190
[CN] tetrachyrin
[NS] *Tetrachyrin orizabaenis*
[REF] 97



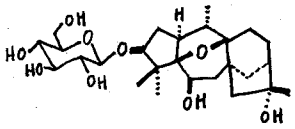
191
[CN] effusin
[NS] *Rabdosia effusus*
[REF] 98



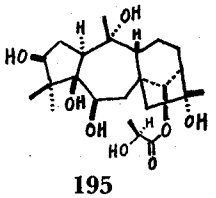
192
[CN] grayanotoxin II
[REF] 99



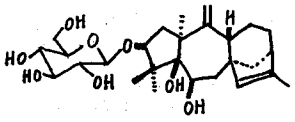
193
[CN] grayanoside C
[NS] *Leucothoe grayana*
[REF] 100



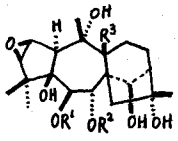
194
[CN] grayanoside D
[NS] *Leucothoe grayana*
[REF] 101



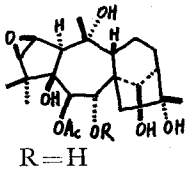
195
[CN] asebotoxin-X
[NS] *Pieris japonica*
[REF] 102, 103



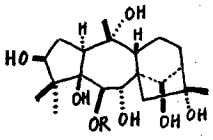
196
[CN] pieroside A
[NS] *Pieris japonica*
[REF] 102



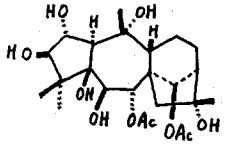
197 R^1=R^2=R^3=H
198 R^1=Ac, R^2=H, R^3=OH
199 R^1=Ac, R^2=COC_2H_5, R^3=OH
[CN] pieristorin H, J, K
[NS] *Pieris japonica*
[REF] 103



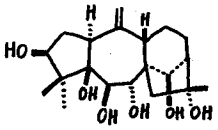
200 R=H
201 R=Ac
[CN] kalmitoxin IV, V
[NS] *kalmia latifolia*
[REF] 104



202 R=H
203 R=Ac
[CN] kalmitoxin I, III
[NS] *Kalmia latifolia*
[REF] 104



204
[CN] kalmitoxin VI
[NS] *Kalmia latifolia*
[REF] 104



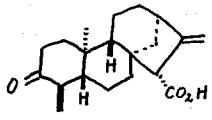
205
[CN] kalmitoxin II
[NS] *Kalmia latifolia*
[REF] 104

Additional references

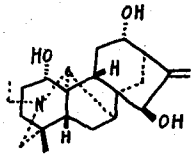
[REF] 105
[NC] studies of constituents of *Sideritis flavoviren*

[REF] 106
[NC] studies of constituents of *Eupatorium tinofolium*

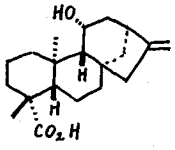
2) Syntehsis and Reaction



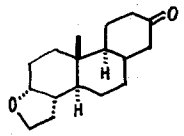
206
[REF] 107
[NC] synthesis of optically active **206**



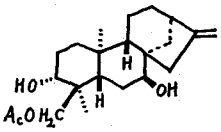
207
[CN] napelline
[REF] 108
[NC] total synthesis



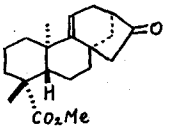
208
[REF] 109
[NC] conversion of **165** to **208** and 12-hydroxy derivatives



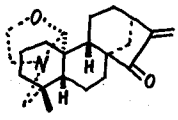
209
[REF] 110
[NC] intermediate to epoxynorcafestanone



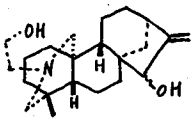
210
[REF] 111
[NC] oxidation with SeO_2–H_2O_2



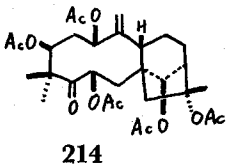
211
[REF] 112
[NC] 10α→9α-methyl migration



212
[REF] 113
[NC] alumina catalyzed addition of amine to **212**

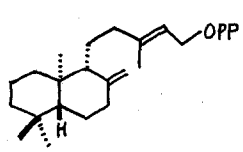


213
[REF] 114
[NC] oxidative cyclization



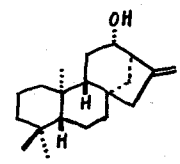
214
[REF] 115
[NC] conversion of grayanotoxin II tetraacetate to **214**

3) Miscellaneous Section



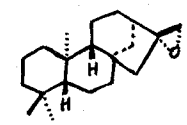
215

[CN] copalyl pyrophosphate
[REF] 116, 117
[NC] enzymatic cyclization of **215** to *ent*-kaurene



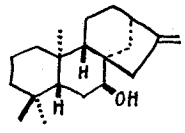
216

[REF] 118
[NC] microbial transformation of **216**



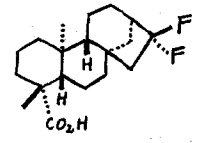
217

[REF] 119
[NC] inhibition of gibberellic acid biosynthesis by **217**



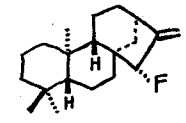
218

[REF] 120
[NC] microbial transformation of diterpenoids



219

[REF] 121
[NC] microbial transformation



220

[REF] 122
[NC] Biological activity of some fluoroditerpenoids

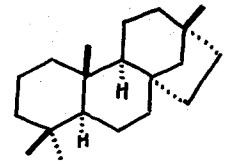
Additional references

[REF] 123, 124
[NC] ^{13}C NMR studies

[REF] 125
[NC] analytical studies of stevioside of *stevia rebaudiana*

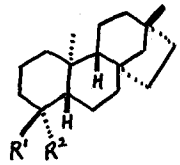
[REF] 126
[NC] A review article on diterpenoid alkaloids

X. BEYERANE DERIVATIVES

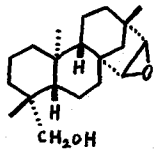


Beyerane

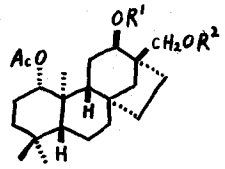
1) Isolation and Structure Determination



221 R^1=Me, R^2=CH_2OH
[CN] erythroxylol-A
222 R^1=CH_2OH, R^2=Me
[NS] *Baccharis tola*
[REF] 20

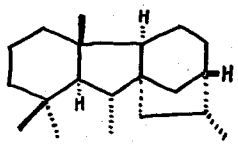


223
[NS] *Baccharis tola*
[REF] 20



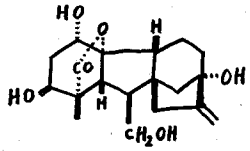
224 R^1=H, R^2=Ac
225 R^1=Ac, R^2=H
[NS] *Sideritis serrata*
[REF] 127

XI. GIBBERELLANE DERIVATIVES



Gibberellane

1) Isolation and Structure Determination



226
[CN] gibberellin A_{57}
[NS] *Gibberilla fujikuroi*
[REF] 128

Additional references

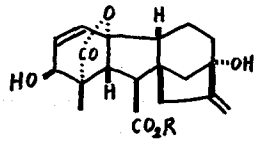
[REF] 129
[NC] identification of gibberellins in Spinach shoots

[REF] 130
[NC] identification of gibberellins in wheat grain

[REF] 131
[NC] gibberellins in mature apple seeds
[NS] *pharbitis purpurea*

[REF] 132
[NC] glucosyl esters of GA_5 and A_{44}

2) Synthesis and Reaction

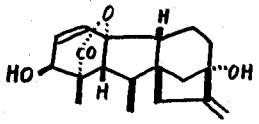


227 R=H

[CN] gibberellin A_3
[REF] 133, 134
[NC] total synthesis
[REF] 135
[NC] introduction of fluorine at C–4
[REF] 136
[NC] rearrangement
[REF] 137
[NC] Pd $(OAc)_2$-catalyzed reactions
[REF] 138
[NC] partial synthesis of 16, 17-dihydro-Δ^{15}-G A_3

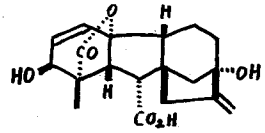
228 R=CH_3

[REF] 139
[NC] photochemistry, crystal structure
[REF] 140
[NC] introduction of chlorine and bromine at C–7
[REF] 141
[NC] 3-O-acetyl derivative, reaction with PBr_3



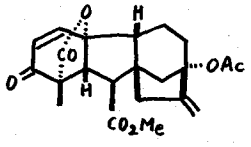
229

[REF] 142
[NC] chemical conversion from GA_3



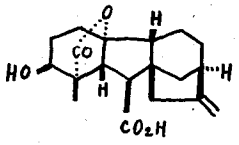
230

[CN] 6-*epi*-gibberellin A_3
[REF] 143
[NC] chemical conversion from GA_4



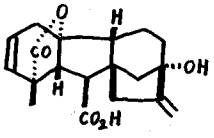
231

[REF] 144
[NC] reduction with $NaBH_4$ or $NaBD_4$



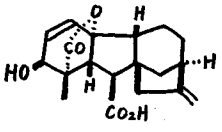
232

[CN] gibbreellin A_4
[REF] 145
[NC] total synthesis
[REF] 146
[NC] physiological studies
[REF] 147
[NC] deuterium labeling



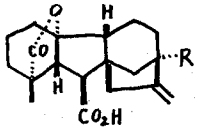
233

[CN] gibberellin A_5
[REF] 148
[NC] 1-deuterated derivative
[REF] 149
[NC] partial synthesis, 3-deuterated derivatives



234

[CN] gibberellin A_7
[REF] 150
[NC] partial synthesis



235 R=H

[CN] gibberellin A_9

[REF] 147

[NC] deuterium labeling

[REF] 149

[NC] partial synthesis, 3-deuterated derivative

236 R=OH

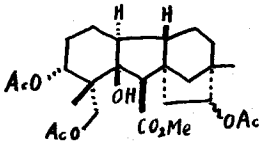
[CN] gibberellin A_{20}

[REF] 147

[NC] deuterium labeling

[REF] 149

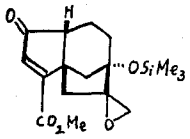
[NC] partial synthesis 3-deuterated derivative



237

[REF] 151

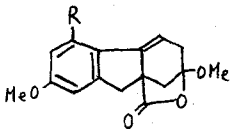
[NC] synthesis



238

[REF] 152

[NC] synthesis

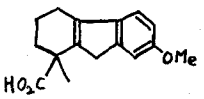


239 R=H

240 R=OMe

[REF] 153

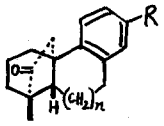
[NC] synthesis



241

[REF] 154

[NC] synthesis

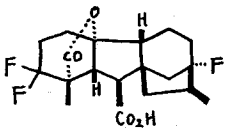


242 n=1 or 2, R=H

243 n=1or 2, R=OMe

[REF] 155

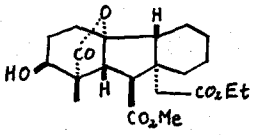
[NC] synthesis



244

[REF] 156

[NC] chemical conversion from benzyl gibberellate

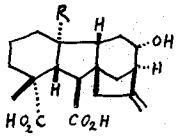


245

[REF] 157

[NC] photoproduct from gibberellin C, X-ray analysis

3) Miscellaneous Section

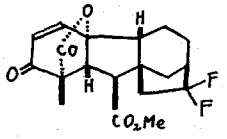


250 R=Me

251 $R=CO_2H$

[REF] 118

[NC] microbial transformations



252

[REF] 121

[NC] microbial transformations

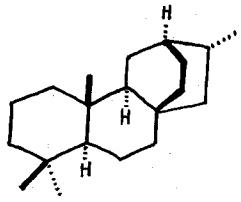
Additional references

[REF] 158
[NC] application of gibberellic acid in treatment of lung cancer
[REF] 159
[NC] phytohormone activity of gibberellin A_3

[REF] 160
[NC] HPLC separation of gibberellins
[REF] 161
[NC] mass spectrometric studies

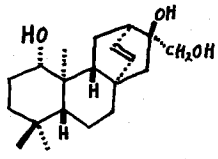
[REF] 162
[NC] a review on plant hormone
[REF] 163
[NC] metabolism of gibberellins

XII. ATISANE DERIVATIVES



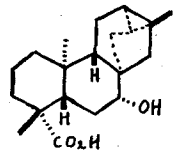
Atisane

1) Isolation and Structure Determination



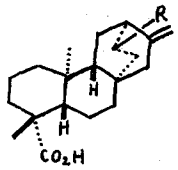
253

[CN] sideritol
[REF] 164
[NC] X-ray analysis of *p*-bromobenzoate

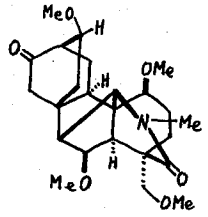


254

[CN] ciliaric acid
[NS] *Helianthus niveus*
[REF] 85

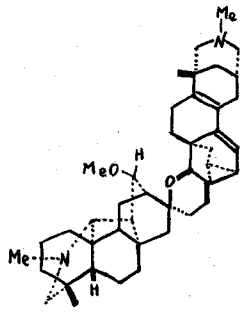


255 R=O-ang
256 R=O-isoval
[NS] *Helianthus* species
[REF] 3



257

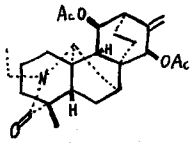
[REF] 165
[NS] X-ray analysis, intermediate for total synthesis



258

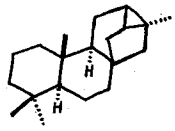
[CN] staphisine
[NS] *Delphinium staphisagria*
[REF] 166
[NC] X-ray analysis

2) Synthesis and Reaction



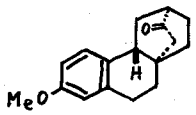
259

[REF] 167
[NC] total synthesis



260

[REF] 168
[NC] chemical conversion



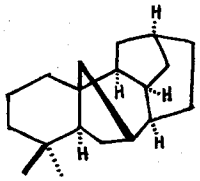
261

[REF] 169
[NC] synthesis

3) Miscellaneous Section

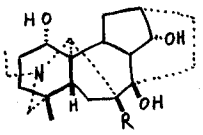
[REF] 41
[NC] a review
[REF] 126
[NC] a review

XIII. ACONANE DERIVATIVES

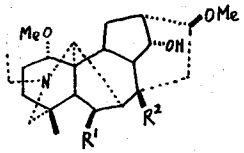


Aconane

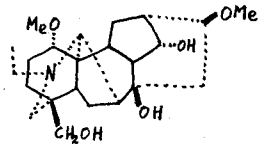
1) Isolation and Structure Determination



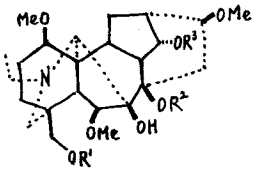
262 R=H
[CN] cardiopetaline
263 R=OH
[CN] cardiopetalidine
[NS] *Delphinium cardiopetalum*
[REF] 170
[NC] X-ray analysis of **262**



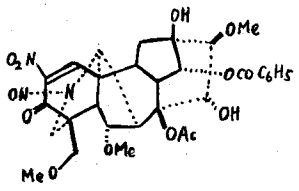
264 R^1=OAc, R^2=OMe
265 R^1=R^2=OH
[NS] *Delphinium bicolor*
[REF] 171
[NC] X-ray analysis



266
[CN] cammaconine
[NS] *Aconitum variegatum*
[REF] 172
[NC] structure revision



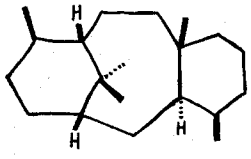
267 R^1=R^2=Me, R^3=Ac
[CN] ambiguine
268 R^1=Me, R^2=H, R^3=Ac
[CN] 14-acetylbrowniine
269 R^1=COC_6H_4-*o*-NHAc, R^2=H, R^3=Ac
[CN] ajadine
270 R^1=COC_6H_4-*o*-N(Me)C(=O)CH$_2$C(=O), R^2=H, R^3=Bz
[CN] ajacusine
[NS] *Consolida ambigua*
[REF] 173



271
[REF] 174
[NC] oxidation product of aconitine

Additional reference
[REF] 126
[NC] a review

XIV. TAXANE DERIVATIVES

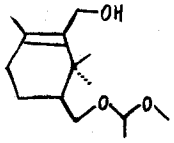


Taxane

1) Isolation and Structure Determination

[REF] 175
[NC] a review

2) Synthesis and Reaction

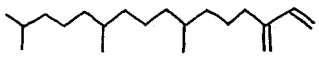


272

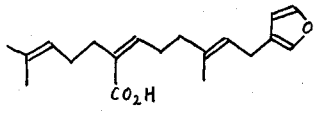
[REF] 176
[NC] synthesis of a key intermediate

XV. THE OTHERS

1) Isolation and Structure Determination

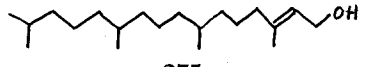


273



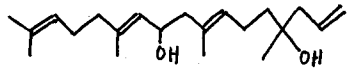
274

[CN] neophytadiene (**273**)
centipedic acid (**274**)
[NS] *Plagiocheilus prostratus*
[REF] 177

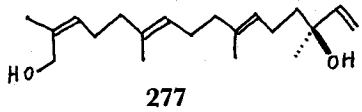


275

[CN] phytol
[NS] *Thymelea hirsuta*
[REF] 178

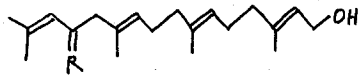


276

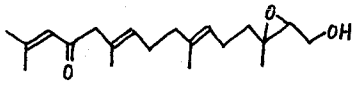


277

[NS] *Nicotiana sylvestris*
[REF] 179

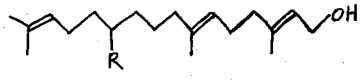


278 R=O **279** R=OH, H

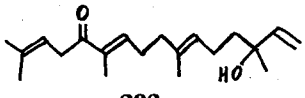


280

[CN] eleganolone (**278**), elegandiol (**279**)
epoxy eleganolone (**280**)
[NS] *Cystoseira elegans* and *Bifurcaria bifurcata*
[REF] 180 and 181

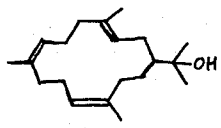


281 R=CH_2OH
282 R=CHO
[NS] *Croton kerrii*
[REF] 182



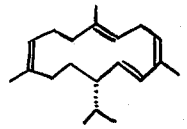
283

[REF] 180
[NC] acyclic diterpenes from mediterranen cystoseiraceae



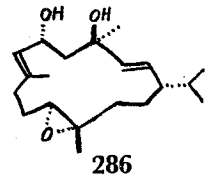
284

[NS] *Helichrysum* species
[REF] 4



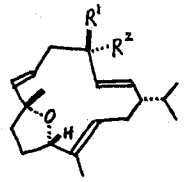
285

[CN] cembrene
[NS] *Nicotiana tabacum*
[REF] 183

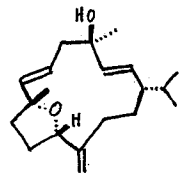


286

[NS] Greek tobacco
[REF] 184

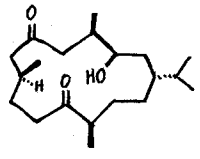


287 R^1=OH, R^2=Me
288 R^1=Me, R^2=OH



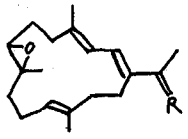
289

[NS] Greek tobacco
[REF] 184

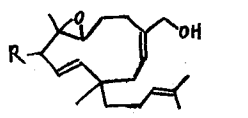


290

[CN] plexaurolone
[NS] *Plexaura*-related species
[REF] 185
[NC] X-ray analysis

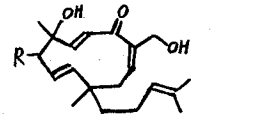


291 R=Me, H
292 R=CH_2
[NS] *Sarcophyton crassocaule*
[REF] 186



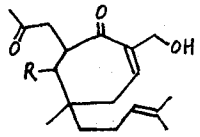
293 R=OC–CH=
O

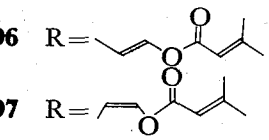
294 R=H



295 R=O–C–CH=
O

[CN] vibsamine A (**293**)
vibasamine B (**295**)
vibsamine F (**294**)
[NS] *Viburnum odoratissimum*
[REF] 187

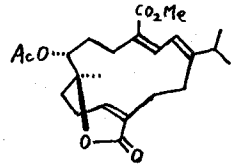




296 R=

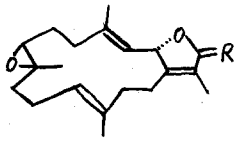
297 R=

[CN] vibsamine C (**296**)
vibsamine D (**297**)
[NS] *Viburnum odoratissimum*
[REF] 187

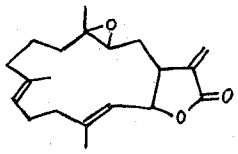


298

[CN] emblide
[NS] *Sarcophyton glaucum*
[REF] 188
[NC] X-ray analysis

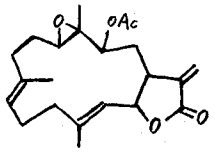


299 R=H_2
300 R=O
[CN] deoxosarcophine (**299**)
sarcophine (**300**)
[NS] *Sarcophyton* species
[REF] 189

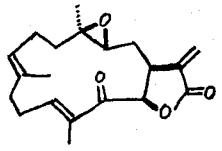


301

[NS] *Lobophytum pauciflorum*
[REF] 190

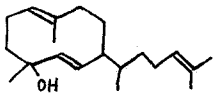


302

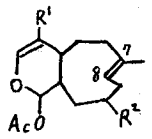


303

[CN] peunicin
[NS] *Eunicea succinea*
[REF] 191
[NC] X-ray analysis



304

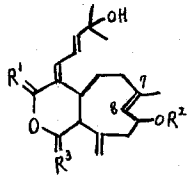


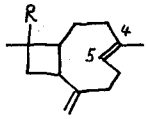
[CN] obscuronatin (**304**), xeniculin (**305**)
[NS] *Xenia macrospiculata and X. obscuronate*
[REF] 192

305 R^1=  , R^2=OAc

306 R^1=  , R^2=H

307 R^1=  , R^2=H, 7, 8-epoxide

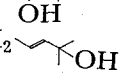


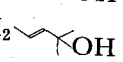


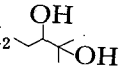
308 R^1=O, R^2=H, R^3=H_2
309 R^1=H_2, R^2=H, R^3=O
310 R^1=H_2, R^2=Ac, R^3=O
311 R^1=O, R^2=H, R^3=H_2, 7, 8-epoxide
312 R^1=H_2, R^2=H, R^3=OH, H

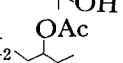
313 R=CH_2 

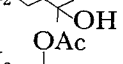
314 R=CH_2  , 4.5-epoxide

315 R=CH_2 

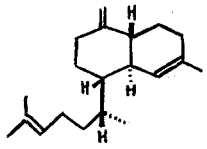
316 R=CH_2  , 4.5-epoxide

317 R=CH_2 

318 R=CH_2 

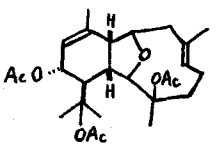
319 R=CH_2  , 4.5-epoxide

[CN] xeniolide-A (**308**) xeniolide-B (**309**), xenialactol (**312**), xeniaphyllenol (**313**), isoxeniaphyllenol (**315**)
[NS] *Xenia macrospiculata* and *X. obscuronate*
[REF] 192



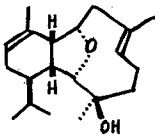
320

[NS] *Cubitermes umbratus*
[REF] 193
[NC] X-ray analysis



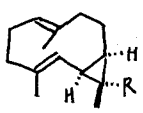
321

[CN] ophirin
[NS] *Muricella* spesies
[REF] 194

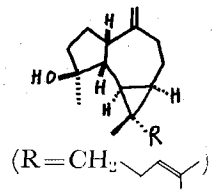


322

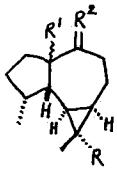
[NS] pacific soft coral
[REF] 195



323

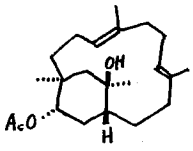


324



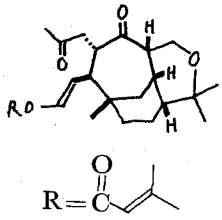
325 $R^1=\beta$–H, $R^2=CH_2$
326 $R^1=\alpha$–H, $R^2=CH_2$
327 $R^1=\alpha$–H, $R^2=\alpha$–OH, β–H

[CN] cneorubin Y (**323**), cneorubin X (**324**)
cneorubin W_1 (**325**), cneorubin W_2 (**326**)
cneorubin U (**327**)
[NS] *Cneorum tricoccon* and *Neochamaelea pulverulenta*
[REF] 196



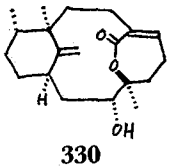
328

[CN] secotrinervitane
[NS] *Nasutitermes princeps*
[REF] 197
[NC] X-ray analysis



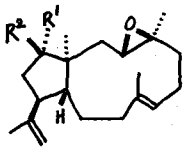
329

[CN] vibsamin E
[NS] *Viburnum odoratissium*
[REF] 187 and 198
[NC] X-ray analysis

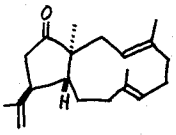


330

[CN] cleomeolide
[NS] *Cleome viscosa*
[REF] 199
[NC] X-ray analysis

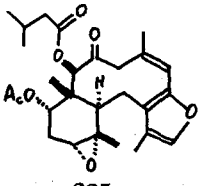


331 R_1, R_2=O,
332 R_1=OH, R_2=H
333 $R_1=R_2$=H

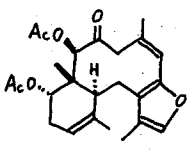


334

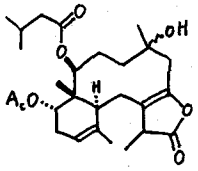
[NS] *Dictyota dichotoma*
[REF] 200



335

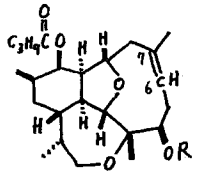


336

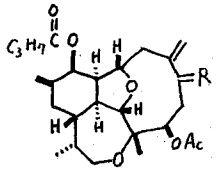


337

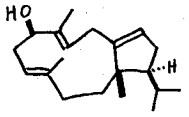
[NS] *Scytalium tentaculatum*
[REF] 201



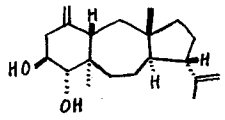
338 R=Ac, 6, 7-trans
[CN] asbestinin-1
339 R=H, 6, 7-trans
[CN] asbestinin-3
340 R=Ac, 6, 7-cis
[CN] asbestinin-2
[REF] 202
[NC] X-ray analysis (**338**)



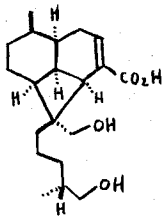
341 R=O
[CN] asbestinin-4
342 R=α-OH, β-H
[CN] asbestinin-5



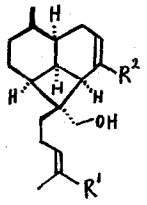
343



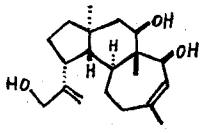
344
[NS] *Clavularia inflata*
[REF] 203



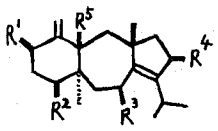
345
[NS] *Eremophila* species
[REF] 204



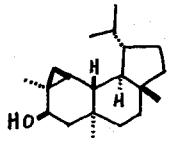
346 $R^1=R^2=CH_2OH$
347 $R^1=CO_2H$, $R^2=Me$
348 $R^1=CO_2H$, $R^2=CH_2OH$
[NS] *Eremophila decipiens*
[REF] 205



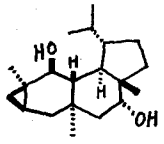
349
[CN] onychiol B
[NS] *Onchium japonicum*
[REF] 206
[NC] X-ray analysis



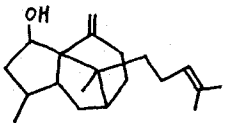
350 R^1=H, $R^2=R^5$=OH, $R^3=R^4$=OAc
351 $R^1=R^3=R^2$=H, $R^2=R^5$=OH
352 $R^1=R^5$=OH, $R^2=R^3=R^4$=H
353 $R^1=R^3=R^4=R^5$=H, R^2=OH
[CN] amijidictyol (**350**), amijiol (**351**), isoamijiol (**352**)
[NS] *Dictyota linearis*
[REF] 207 and 208



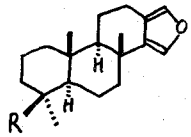
354
[CN] neoverrucosan-5β-ol
[NS] *Mylia verrucosa*
[REF] 209
[NC] X-ray analysis



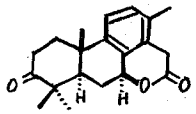
355
[CN] dihydroxy-verrucosane
[NS] *Mylia verrucosa*
[REF] 210
[NC] X-ray analysis



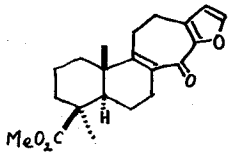
356
[CN] stoechospermol
[NS] *Stoechospermum marginatum*
[REF] 211



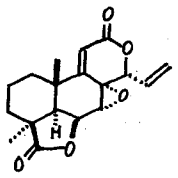
357 R=CO_2H
358 R=CHO
359 R=Me
[NS] *Spongia officinalis*
[REF] 212



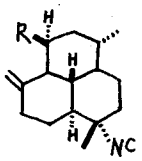
360
[NS] *Vellozia compacta*
[REF] 213



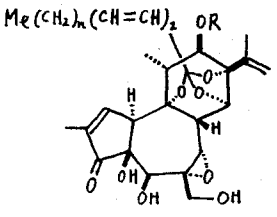
361
[CN] hispanonic acid methyl ester
[NS] *Balloya hispanica*
[REF] 214
[NC] X-ray analysis



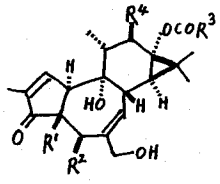
362
[CN] Salignone-D
[NS] *Podocarpus saligna*
[REF] 215
[NC] X-ray analysis



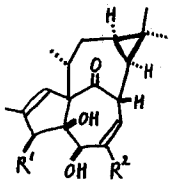
363 R=CH=CMe_2
364 R=CH_2— 
365 R=CH_3—  NC
[NS] *Adosia* species
[REF] 216



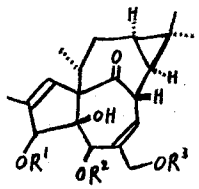
R=$CO(CH=CH)_3(CH_2)_2CH_2OY$
n=2
366 Y=H, **367** Y=$CO(CH_2)_{12}Me$
368 Y=$COCH=CH(CH_2)_{11}Me$
369 Y=$COC_{13}H_{27}$
370 Y=$COC_{14}H_{27}$
[NS] *Stillingia sylvatica* [REF] 217



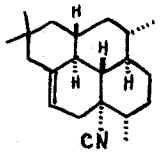
371 R^1=R^2=H, R^3=$CH_2CH\ Me_2$
R^4=OCOC(Me)=CHMe
[NS] *Synadenium grantii*
[REF] 218
372 R^1=R^2=OH, R^4=H
R^3=$(CH_2)_{14}Me$
[NS] *Stillingia sylvatica*
[REF] 217



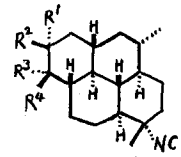
373 R^1=angelate, R^2=Me
374 R^1=hexanoate, R^2=Me
375 R^1=angelate, R^2=CH_2OH
[NS] *Euphorbia paralias*
[REF] 219
[NC] Irritant and cytotxic constituents



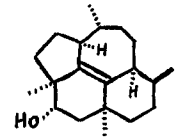
376 $R^1=COC_2H_6$, $R^2=H$, $R^3=COCH(Me)C_2H_5$
377 $R^1=R^2=H$, $R^3=COCHMe_2$
378 $R^1=COC_2H_5$, $R^2=H$, $R^3=COCHMe_2$
399 $R^1=R^3=COCHMe_2$, $R^2=H$
[NS] *Euphorbia cotinifolia* [REF] 220



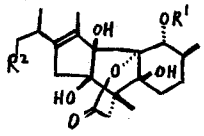
380
[NS] *Adocia* species



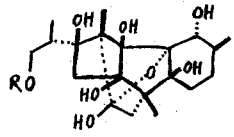
381 $R^1=NC$, $R^2=R^3=Me$, $R^4=H$
382 $R^1=R^4=Me$, $R^2=H$, $R^3=NC$
[REF] 216



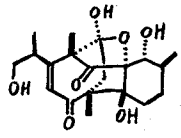
383
[NS] *Nasutitermes rippertii* and *N. ephratae*
[REF] 221
[NC] X-ray analysis



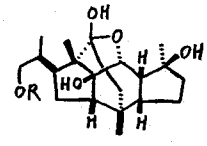
384 $R^1=Ac$, $R^2=H$
385 $R^1=R^2=H$
386 $R^1=H$, $R^2=OH$
387 $R^1=H$, $R^2=O$-β-p-glc·pyr



388 R=H
389 R=β-D-glc·pyr



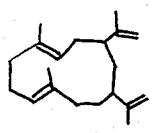
390



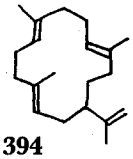
391 R=H
392 R=β-D-glc·pyr

[CN] cinncassiol-A (**386**), -B (**388**), -C (**390**), -D (**391**)
[NS] *Cinnanomum cassia*
[REF] 222, 223, 224, 225
[NC] X-ray analysis

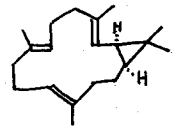
2) Synthesis and Reaction



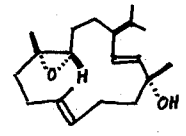
393
[CN] (±)-cubitene
[REF] 226
[NC] ·intramolecular coupling of allylic dibromide
·total synthesis



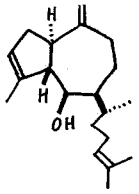
394
[CN] neocembrene
[REF] 227
[NC] ·synthesis of d- and l-neocembrenes and their geometrical isomers
·pheromone activity



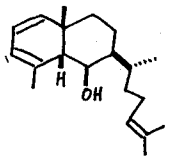
395
[CN] (−)-casbene
[REF] 228
[NC] total synthesis from 1R, 3S(+)-cis-chrystanthemic acid



396
[CN] isocembrol (thunbergol)
[REF] 229
[NC] epoxidation

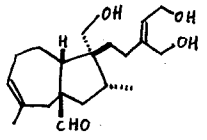


397



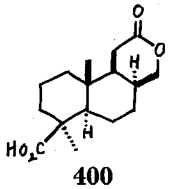
398

[CN] (+)-pachydictyol A (**397**)
(−)-dictyolene (**398**)
[REF] 230
[NC] total syntheses from
(−)-α-santonin



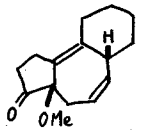
399

[CN] (±)-portulal
[REF] 231
[NC] total synthesis
[REF] 232
[NC] selective degradation of side chain



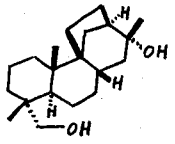
400

[CN] dihydro-8-epi-acrostalidic acid
[REF] 233
[NC] total synthesis



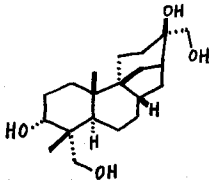
401

[REF] 234
[NC] ·synthetic approach to phorbol
·divinylcyclopropane rearrangement



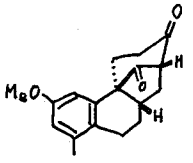
402

[CN] (±)-stemarin
[REF] 235
[NC] total synthesis



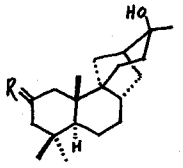
403

[CN] (±)-aphidicolin
[REF] 236
[NC] total synthesis

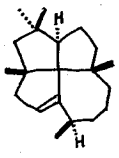


404

[REF] 237
[NC] ·synthetic approach to aphidicolin
·thermolysis of benzocyclobutene

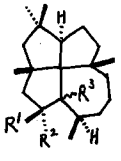


405 R=α-OH, β-H
406 R=O
[CN] (±)-stemodin (**405**)
(±)-stemodinone (**406**)
[REF] 238
[NC] total synthesis

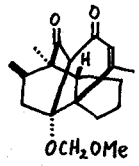


407

[CN] lauren-1-ene
[REF] 239
[NC] ·osmic acid oxidation
·NMR spectra



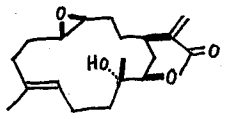
408 R^1=OH, R^2=H, R^3=α-H
409 R^1=H, R^2=OH, R^3=α-H
410 R^1=OH, R^2=H, R^3=β-H
411 R^1=H, R^2=OH, R^3=β-H
[REF] 240
[NC] remote functionalization with $I_2/Pb(OAc)_4$



412

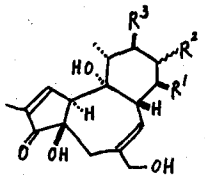
[REF] 241
[NC] ·synthetic precursors to pleuromutilin
·one-step synthesis

3) Miscellaneous Section



413

[CN] flexibilide
[REF] 242
[NC] ·^{13}C-NMR study
·spin-lattice
relaxation measurements



414

[REF] 243
[NC] ·structure-activity relations for irritant and tumor promoting activity
·tigliane derivatives (**414**)
R^1=H or C(Me)=CH_2
R^2=α- or β-$OOC(CH_2)_{12}Me$
R^3=H or $OOC(CH_2)_6Me$

Additional references

[REF] 244
[NC] structure-activity relationships for phorbol-related diterpene esters
[REF] 41 [NC] a review article on the chemistry of the *Composites* (cembrene type diterpenoids)
[REF] 81 [NC] a review article on the plants and plants products that induced contact dermatitis (tigliane, daphnane, ingenane type diterpenoids)
[REF] 245 [NC] a review article on the drugs from marine organisms (cembranoid)

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